BIBLIOGRAPHIC DATA SHEET	1. Report No. AP-103	2. PB 210 736

4. Title and Subtitle
Paint Technology and Air Pollution: A Survey and Economic Assessment

5. Report Date February 1972

6.

7. Author(s)
J. W. Spence and F. H. Haynie

8. Performing Organization Rept. No.

9. Performing Organization Name and Address
ENVIRONMENTAL PROTECTION AGENCY
Office of Air Programs
National Environmental Research Center
Research Triangle Park, North Carolina

10. Project/Task/Work Unit No.

11. Contract/Grant No.

12. Sponsoring Organization Name and Address
Same

13. Type of Report & Period Covered

14.

15. Supplementary Notes

16. Abstracts The objectives were: (1) to survey the technical developments occurring within the paint industry, (2) to identify the characteristics of pollutant attacks on exterior paints, and (3) to estimate the annual cost of air pollutant damage to such paints. New paint formulations and new application techniques are emerging within the paint industry. Factory-applied coatings (industrial finishes) appear to have a bright future in the 1970's. Latex or water-base paints have captured substantially more than 50 percent of the household painting market and are spilling over into the industrial market. The chemical attack of certain air pollutants on exterior finishes is reviewed. Hydrogen sulfide, which is a localized pollutant, is known to attack in-service exterior house paints. In laboratory studies ozone and sulfur dioxide have also been shown to damage paints. There is evidence as well that sulfur dioxide attacks certain pigments of exterior paints. Dose-response data for these pollutants are lacking. An economic assessment was made of the chemical damage of air pollutants on four classes of exterior paints: (1) household, (2) automotive refinishing, (3) coil coating, and (4) maintenance. The total estimated cost at the consumer level is over $0.7 billion annually. Of the four types of exterior paints, household paint sustains the most damage, representing over 75 percent of the total dollar loss.

17. Key Words and Document Analysis. 17a. Descriptors
Air pollution
Paints
Deterioration
Expenses
Hydrogen sulfide
Ozone
Sulfur dioxide
Buildings
Maintenance
Ground vehicles

17b. Identifiers/Open-Ended Terms

17c. COSATI Field/Group

18. Availability Statement Superintendent of Documents
U. S. Goverment Printing Office
Washington, D. C. 20402
SOD: EP 4.9:103 Price $0.35

19. Security Class (This Report) UNCLASSIFIED

20. Security Class (This Page UNCLASSIFIED

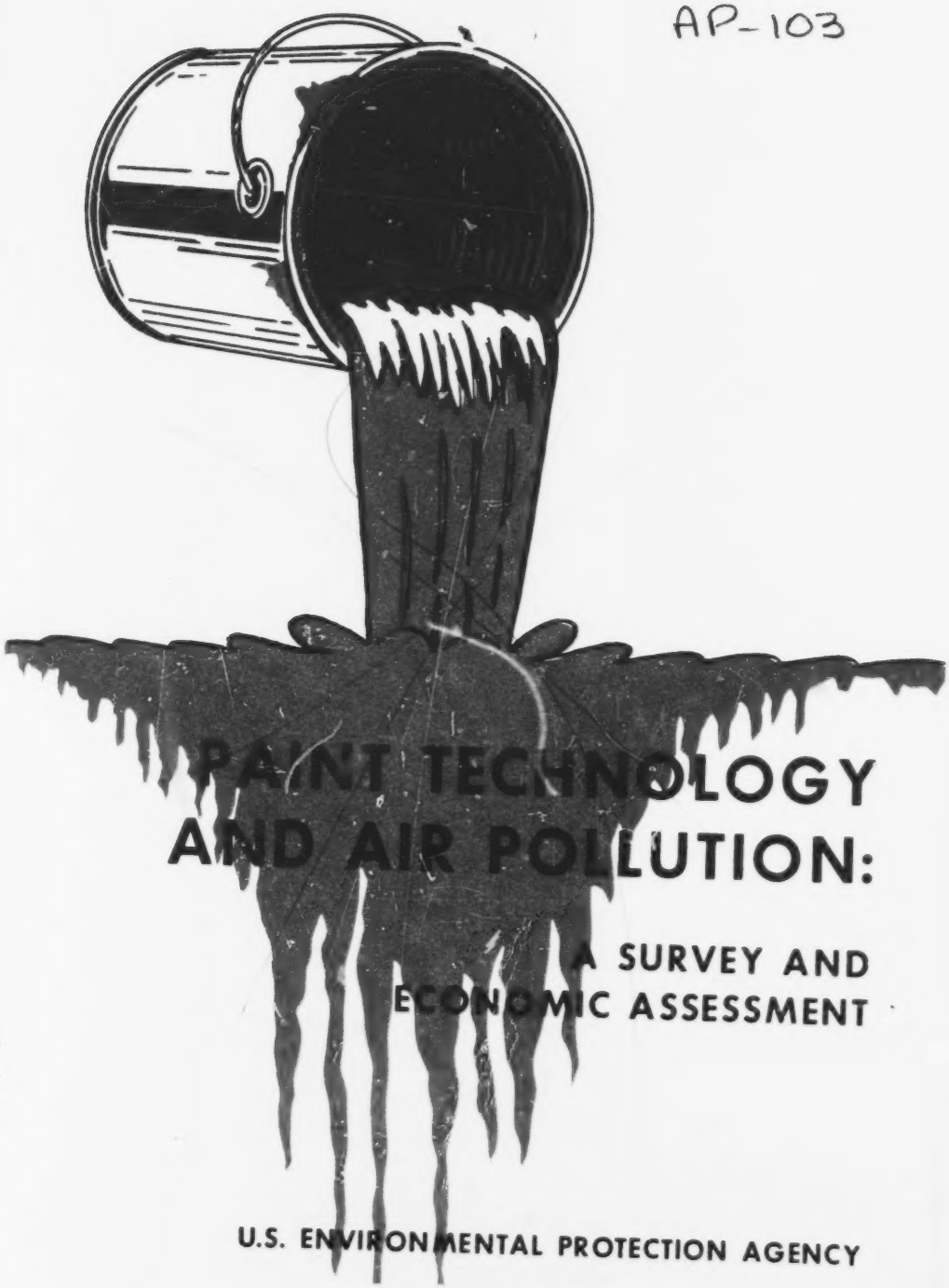
21. No. of Pages 49

22. Price $2.35

FORM NTIS-35 (10-70)

USCOMM-DC 40329-P71

AP-103



PAINT TECHNOLOGY AND AIR POLLUTION:

A SURVEY AND ECONOMIC ASSESSMENT

U.S. ENVIRONMENTAL PROTECTION AGENCY

Reproduced by
NATIONAL TECHNICAL
INFORMATION SERVICE
U.S. Department of Commerce
Springfield, VA 22151

U.S.

49

PAINT TECHNOLOGY AND AIR POLLUTION: A SURVEY AND ECONOMIC ASSESSMENT

J.W. Spence
and
F.H. Haynie
National Environmental Research Center

ENVIRONMENTAL PROTECTION AGENCY
Office of Air Programs
Research Triangle Park, North Carolina
February 1972

[redacted]

Office of Air Programs Publication No. AP-103

ACKNOWLEDGMENT

The authors wish to thank B. G. Brand, Senior Research Chemist, Battelle Memorial Institute, for technical suggestions of value in preparing this manuscript.

CONTENTS

	Page
LIST OF FIGURES	vi
LIST OF TABLES	vii
ABSTRACT	viii
INTRODUCTION	1
DEVELOPMENTS WITHIN THE PAINT INDUSTRY	3
Historical Developments	3
Paint Formulation	7
Binders	7
Pigments	8
Water-Base Paints (Latex)	9
Application Techniques	11
Electrodeposition	11
Coil Coating	12
Electron-Beam Curing	14
DETERIORATION OF EXTERIOR PAINT FILMS	17
Industrial Testing	17
Effects of Air Pollution on Exterior Paints	22
Sulfur Dioxide	24
Hydrogen Sulfide	25
Particulate Matter	26
ECONOMIC ASSESSMENT OF AIR POLLUTION DAMAGE TO EXTERIOR PAINTS	31
AREAS FOR FUTURE INVESTIGATION	35
CONCLUSIONS	37
REFERENCES	39
BIBLIOGRAPHY	43

Preceding page blank

LIST OF FIGURES

Figure *Page*

1 Initial Creep of EM 7339 Second-Quality Emulsion-Base Paint, Before and After 505-hr Exposure to Ozone at 60°C 21

2 Initial Creep of EM 7338 First-Quality Emulsion-Base Paint, Before and After 505-hr Exposure to Ozone at 60°C 21

3 Relationship of Maintenance Frequency for Exterior Repainting to Particulate Concentration 28

LIST OF TABLES

Table		*Page*
1	History of Coatings Industry in the United States	4
2	1968 Trade and Industrial Paint Sales	5
3	U. S. Shipments of Paints, Varnishes, and Lacquers	6
4	Consumption of Resins by Coatings Industry, 1968 to 1975	8
5	Pigment Consumed by Paint Industry, 1968 to 1975	9
6	Estimated Trade Sales of Latex Paints	10
7	Shipment of Coil-Coated Steel and Aluminum	13
8	Coil-Coated Organic Resins	14
9	Types of Film Failure	18
10	ASTM Test Methods for Exterior Paint Films	19
11	Durability of Organic Resin Topcoats	20
12	Mean Concentrations of Three Atmospheric Pollutants 1967	22
13	Relative Pollution Damage to Materials and Availability of Useful Information	23
14	Interval for Exterior Repainting as Function of Particulate Concentration in Five U. S. Cities	27
15	Mean Annual Frequency for Exterior Wall Painting in Philadelphia Area as a Function of Particulate Concentration	29
16	Economic Assessment of Chemical Deterioration of Exterior Paints	32

ABSTRACT

The objectives of this study are: (1) to survey the technical developments occurring within the paint industry, (2) to identify the characteristics of pollutant attacks on exterior paints, and (3) to estimate the annual cost of air pollutant damage to such paints.

New paint formulations and new application techniques are emerging within the paint industry. Factory-applied coatings (industrial finishes) appear to have a bright future in the 1970's. Latex or water-base paints have captured substantially more than 50 percent of the household painting market and are spilling over into the industrial market.

The chemical attack of certain air pollutants on exterior finishes is reviewed. Hydrogen sulfide, which is a localized pollutant, is known to attack in-service exterior house paints. In laboratory studies ozone and sulfur dioxide have also been shown to damage paints. There is evidence as well that sulfur dioxide attacks certain pigments of exterior paints. Dose-response data for these pollutants are lacking.

An economic assessment was made of the chemical damage of air pollutants on four classes of exterior paints: (1) household, (2) automotive refinishing, (3) coil coating, and (4) maintenance. The total estimated cost at the consumer level is over $0.7 billion annually. Of the four types of exterior paints, household paint sustains the most damage, representing over 75 percent of the total dollar loss.

Key Words: material deterioration, exterior paints, pollutants, economic losses, area surveys, costs.

PAINT TECHNOLOGY AND AIR POLLUTION: A SURVEY AND ECONOMIC ASSESSMENT

INTRODUCTION

The purpose of this study is to provide the Environmental Protection Agency and other scientific and lay readers with technical information pertaining to the effects of air pollutants on exterior paints. The specific goals of this report are: (1) to survey the technical developments within the paint industry; (2) to identify the characteristics of pollutant attacks on exterior paints; and (3) to estimate the annual cost of pollution damage to these paints. This information will be used to set priorities in planning laboratory investigations and future contract studies on the effects of air pollutants on exterior paints.

DEVELOPMENTS WITHIN THE PAINT INDUSTRY

The paint industry is currently being confronted with structural materials such as reinforced plastics, stainless steel, and laminated wood that require little or no painting. The sales of such materials are cutting into the paint industrial market. In order to compete, the paint industry is developing thinner finishes with improved durability for metallic and wooden substrates. Paints with new potential applications, such as fire retardants for household use and coatings for plastic materials, are being developed by the industry. This section discusses trends in the use of raw materials, new techniques emerging within the industry, and outside factors, such as the continuing passage of air pollution legislation, which are influencing these trends and techniques.

HISTORICAL DEVELOPMENTS

The paint industry has emerged in the 1970's as a scientific business; just 40 years ago the industry was largely an art. Paint formulation had its beginning with natural resins and minerals; today the industry spends over $1.4 billion annually for various synthetic resins, chemical additives, and containers.[1] A historical glimpse of the paint industry through 1962 is shown in Table 1.[2]

The industry has grown to more than 1,600 companies with about 1,875 plants. Sherwin-Williams is the largest paint manufacturer, with about 10 percent of the total market. About 27 producers of paint account for 57 percent of the total sales, and fewer than 20 companies sell paints nationwide.[1,3] Paint manufacturing is one of the few remaining industries within the United States that consists of small companies that specialize in a limited product line to be marketed within a geographical region.

The paint industry does not invest heavily in the research and development of raw materials. According to the National Paint, Varnish, and Lacquer Association, the paint industry invests only about 2 percent of its sales in research.[3] The industry has been called a parasite because it depends upon the technology generated within the rubber and plastics industry for new resins for paint formulation, although the paint industry does pay for research indirectly through the price of raw materials it purchases from the chemical industry. In many respects this arrangement is advantageous to the small paint producer, because new advances in materials are available to both small and large paint companies.

The paint industry has two distinct markets, trade sales and industrial sales.

Preceding page blank

Table 1. HISTORY OF COATINGS INDUSTRY IN THE UNITED STATES[2]

Year	Event
1804	First white lead production
1815	First varnish production
1865	First water paint patent (U. S. Patent 50,068 to D. P. Flinn)
1867	Production of ready-mixed paints
1910	Casein powder paints put on market
1923	Nitrocellulose lacquer introduced
1924	Titanium dioxide introduced
1924	Modified phenolic resin made commercially available
1927	Alkyd resins made commercially available
1928	Oil-soluble phenolics introduced
1929	Urea-formaldehyde resin used for modification of alkyd resin
1930	Chlorinated rubber produced
1930	Casein paste paints introduced
1930	Molybdate orange introduced
1933	Vinyl copolymer resins introduced
1934	Oil-base emulsion paints put on market
1937	Phthalocyanine blue introduced
1939	Commercial production of dehydrated castor oil
1939	Melanine-formaldehyde resins made available
1939	Ethylcellulose introduced
1939	Polyurethane resins introduced
1944	Silicone resins made commercially available
1948	Latex wall finishes based on styrene-butadiene put on market
1950	Epoxy resins made commercially available
1950	Polyvinyl acetate and acrylic copolymer latices made commercially available
1957	Latex house paints introduced
1962	Electrodeposition of water-base paints used

Trade-sale paints are shelf products that are sold through retail stores to the general public, professional painters, and builders. These products are largely architectural coatings for interior and exterior surfaces of new and existing structures. Industrial sales paints are products that are formulated and sold to other manufacturers for factory application. Coatings for new automotive vehicles, furniture, and appliances are industrial paints. The industrial market also includes maintenance coatings, which are not factory applied but are specially formulated for particular properties, such as chemical resistance, film toughness, and resistance to high temperatures. Industrial plants and equipment require such paints.

A breakdown of the 1968 paint market, according to use of the coatings, is shown in Table 2. The United States shipments of paints, varnishes, and

Table 2. 1968 TRADE AND INDUSTRIAL PAINT SALES[1]

Type of sale	Sales		
	10^6 gal	10^6	$/gal
Trade	424	1,428	
Interior house paints:			
Latex emulsion	145	425	2.93
Oil and alkyd	45	170	3.78
Primer, sealers, etc.	10	30	3.00
Miscellaneous	25	100	-
Exterior house paints:			
Latex emulsion	55	190	
Oil and alkyd	40	155	3.88
Enamels	15	55	3.66
Primers, sealers, etc.	10	35	3.50
Miscellaneous	20	50	-
Automotive refinishing	30	150	5.00
Traffic	25	45	1.80
Other	4	23	-
Industrial	419	1,159	
Automotive, new	55	170	3.09
Marine	20	85	4.25
Railroad, aircraft, etc.	15	40	2.67
Coil coating	20	80	4.00
Prefinished wood	15	40	2.67
Industrial maintenance	45	160	3.56
Machinery and equipment	30	75	2.50
Miscellaneous	219	509	-

lacquers for 1968 were 843 million gallons valued at $2,587 million.[1] Latex or water-base paints have captured about 50 percent of the architectural trade-sales market. Paints produced for the industrial-sales market are primarily oil- or solvent-formulated.

The shipments of the trade and industrial finishes through the last decade and their projected growth into 1975 are shown in Table 3. Except for 1967, the paint industry has maintained a steady growth during the 1960's with a growth rate of 4.7 percent of the total annual dollar volume. In 1967 the impact of high interest rates caused a drop in new construction and in overall paint sales. It should be pointed out that an undetermined portion of this dollar growth has resulted from continued inflation over this period. Thus a better indication of growth of the paint industry is gallons sold. The average

Table 3. U.S. SHIPMENTS OF PAINTS, VARNISHES, AND LACQUERS[1,4]

Year	Trade		Industrial		Total	
	10^6 gal	$10^6	10^6 gal	$10^6	10^6 gal	$10^6
1961	329	$1,038	294	$ 712	623	$1,750
1962	337	1,078	306	755	643	1,833
1963	373	1,125	305	765	678	1,890
1964	395	1,173	330	829	725	2,002
1965	411	1,247	365	922	776	2,169
1966	416	1,312	421	1,052	837	2,364
1967	398	1,329	383	1,019	781	2,348
1968	424	1,428	419	1,159	843	2,587
1969[a]	439	1,480	435	1,213	874	2,693
1975[a]	540	2,050	600	1,860	1,140	3,910

[a] Estimated.

annual growth (gallonage) during the 1960's was projected to be about 3.3 percent.[1]

From 1962 through 1968, the dollar volume of industrial finishes increased at an annual rate of 6.6 percent, compared with 4.4 percent for trade sales. The rate of increase of industrial paint finish sales is likely to remain ahead of the rate of growth of trade sales in the 1970's. Although construction is expected to increase in the early 1970's and improve the sales of trade paints, the demand for prefinished materials of construction will favor factory-applied coatings at the expense of trade sales. The rising cost of labor for on-site painting is another factor that is increasing the trend toward use of precoated materials. The annual dollar sales of industrial finishes as projected into 1975 are not expected to exceed trade sales. The volume (gallonage) of industrial finishes, however, is projected to take a commanding lead over trade sales by 1975.[4]

Air pollution legislation is also affecting developments within the paint industry. Los Angeles County Rule No. 66, one of the legislative acts most significant to the paint industry, recognizes paint as a source of hydrocarbon emissions.[3] It has been estimated by Los Angeles County officials that the application of paint within the county contributes 20 percent of the total organic emissions, or 360 tons of hydrocarbons per day.[1] Certain hydrocarbons that are used as solvents in paint formulation have been found to react photochemically with nitric oxides to form smog. The damaging effects of smog on materials are not completely understood; however, smog does cause eye irritation and probably respiratory illness.

Los Angeles Rule No. 66, which went into effect July 1, 1967, regulates the amount of daily hydrocarbon emissions and applies especially to all industrial-coating manufacturing techniques as well as to the sale and utilization of architectural coatings. Similar legislation is being considered by officials of other cities because photochemical smog is rapidly becoming a problem in cities across the United States. San Francisco has already adopted a law, Regulation No. 3, that is very similar to Los Angeles Rule No. 66.

Through the National Paint, Varnish, and Lacquer Association, the paint industry has financed the construction and operation of a smog chamber at Battelle Memorial Institute, Columbus, Ohio.[5] This chamber work should provide additional data about the organic materials in paints that undergo photochemical reactions and produce eye irritation. Rule No. 66 has focused on the problem of hydrocarbon air pollution within the paint industry. Consequently, it is very likely that new techniques, such as non-heat-curing processes and paint formulations that avoid photochemically reactive solvents, will be forthcoming within the industry. Water-base paints will probably find increasing use in industrial painting processes.

PAINT FORMULATION

In general, the formula for paints – whether the paints be oil- or water-base – consists of three basic components: (1) a binder, which forms the film and consists of natural resins, drying oils, or synthetic polymers; (2) a solvent, which is either water or an organic solvent; and (3) a pigment, which promotes color and improves film properties.[2] When the binder and solvent are considered to be one component, they are referred to as the "vehicle" of the paint formulation. Other ingredients are present, such as wetting agents and fungicides, that aid in the processing, application, and protection of paint.

Oil- and water-base paint films are formed differently. Oil-base paint forms a film through a combination of organic solvent evaporation and the catalytic polymerization of the drying oil. Water-base paints form a continuous film through evaporation of their water content and subsequent coalescence of the synthetic polymer particles.

Binders

A major trend within the paint industry is the increasing formulation of different resins as binders. The paint industry uses the term "resins" to mean both natural and synthetic organic polymers that are used as binders in the formulation of paints. The present and future consumption of resins by the paint industry is shown in Table 4. This table represents the resins that are consumed in both the trade and industrial markets. This report does not elaborate on the numerous formulations and uses of these resins. The alkyd, polyvinyl acetate, and acrylic resins are briefly discussed.

Alkyds are a group of polyester resins that are oil-modified for paint formulations. These resins are the major binders used by the coatings industry.

Their popularity stems from the fact that they are relatively low-cost, easily applied resins and are compatible with other resins. Alkyd paints are used primarily as industrial baking finishes; however, they still command about 50 percent of the exterior household paint market. Although the total volume of alkyds consumed by the industry continues to grow, as shown in Table 4, their share of the industry (gallonage) is declining with the use of more durable synthetic resins.

Table 4. CONSUMPTION OF RESINS BY COATINGS INDUSTRY, 1968 to 1975[6]

Resin	Consumption, 10^6 lb			
	1968	1969	1970	1975
Alkyds	630.0	673.8	695.7	828.0
Polyvinyl acetate	116.0	142.6	156.6	248.5
Urea and melamine	66.0	72.0	76.3	103.5
Epoxy	62.0	72.9	80.4	127.1
Acrylics	57.0	63.6	68.8	101.2
Styrene butadiene	40.0	37.8	37.0	29.7
Phenolics	36.0	38.0	39.1	47.2
Polyester	14.3	10.5	11.6	18.2

The polyvinyl acetates are the second major group of resins and are perhaps the fastest growing class. The consumption of these resins will probably double by the end of the 1970's. Polyvinyl acetate resins, as well as blends of acrylic, nitrocellulose, and hydroxyl-modified vinyl resin, are primarily used as interior decorative and architectural coatings.

Acrylic resins provide coatings with excellent durability and chemical and heat resistance. Resins with such properties are finding use as exterior architectural finishes and in industrial finishes. Thermosetting-thermoplastic lacquers and enamels of acrylic resins have been formulated for new automotive topcoats and other industrial markets.

Pigments

The paint industry annually consumes millions of pounds of inorganic oxides, sulfates, and carbonates for pigmentation of trade and industrial paints. Both natural and synthetic pigments are used by the industry. Pigments are used in paint formulations to obscure the substrate, to impart color for

aesthetic purposes, and to improve durability of the film. Materials such as talcs, clays, and chalks are used to aid processing and to maintain formulation stability. These materials are called extenders, fillers, and, more recently, supplemental pigments.

White films are the most popular for exterior house paints. White pigments are also utilized to formulate most tints and light hues. The quantity of white pigments consumed by the paint industry in the past 3 years and the projected consumption for 1975 are shown in Table 5.

Table 5. PIGMENT CONSUMED BY PAINT INDUSTRY, 1968 to 1975[6]

Pigment	Consumption, 10^6 lb			
	1968	1969	1970	1975
Titanium dioxide	377.0	396.2	412.1	516.0
Zinc oxide	36.0	32.3	29.8	13.2
Zinc dust	25.0	27.4	29.2	41.9
Lead, red, dry	14.2	14.4	14.6	15.8
Lead, white	7.2	6.9	6.3	2.6

White lead pigment was once used exclusively to formulate household paints. Titanium dioxide is now being used increasingly in trade and industrial finishes because of its superior hiding power and color retention. The dwindling use of lead pigments can also be partially attributed to their toxicity.

Water-Base Paints (Latex)

The national concern over air pollution will certainly provide impetus within the paint industry for the further development of water-base paints and solvent-free coatings. Water-base paints or latex house paints probably represent the most significant development in paint formulation since World War II. The popularity and rapid growth in the use of water-base paints as architectural trade coatings have been described as phenomenal. Certainly ease of application and cleanup, as well as good performance, have contributed to the popularity and success of water-base paints among weekend painters and paint contractors.

It is quite evident that natural products have lost a significant portion of the trade sales market to the newer synthetic resins. Linseed oil, the major natural oil used by the paint industry, was once the principle oil used in exterior house paints. The consumption of this oil, 245 million pounds in 1963, was only 150 million pounds in 1968. This decline can be attributed directly to the increased

use of water-base paints for exterior coatings.[1,6] As shown in Table 2, water-base systems have largely captured the architectural interior market. Latex exterior paints have also claimed more than 50 percent of the architectural exterior market.

Projections indicate that latex paints will have about 70 percent of the trade-sale exterior market in 1975. A breakdown of the gallonage of the major latex paints is shown in Table 6.

Table 6. ESTIMATED TRADE SALES OF LATEX PAINTS[1]

Paint	Sales, 10^6 gal			
	1966	1967	1970	1975
Interior paints				
Polyvinyl acetate	45	46	48	53
Vinyl acetate-acrylic	7	10	22	35
Styrene-butadiene	29	26	18	15
Acrylic	12	16	24	35
Exterior paints				
Polyvinyl acetate	11	11	9	7
Vinyl acetate-acrylic	11	16	28	45
Acrylics	21	24	30	44

Acrylic binders and their various blends are predicted to capture the architectural exterior paint market in the early 1970's. Acrylic resins make possible the manufacture of paint with a superior film stability that will withstand both ultraviolet light and weathering, two factors that must be overcome for exterior durability. Straight acrylic resins are considerably higher in price than other resins. This disadvantage can be overcome by copolymerization with cheaper monomers, although this procedure does sacrifice some film properties.

Water-base paints are just beginning to emerge within the industrial market. Their adaptation to industrial application has been much slower than predicted for two reasons: cost and formulation problems. Most of the industrial coatings applied to consumer items require high-performance finishes with high gloss. These properties have been difficult to obtain with water-formulated paints,

but semi-gloss latex paints are emerging on the market. As yet, however, their properties do not compare with those of oil-base enamels. In addition, water is more costly to evaporate from baking finishes than organic solvents, and the rate of evaporation is difficult to control, which means that the industry must face the cost of revamping equipment.

Nevertheless, several important reasons exist for eliminating organic solvents from paint formulations. From a safety point of view, the use of water-base systems minimizes the chance of fire and may make possible a reduction in insurance premiums. Also, the solvent fumes, consisting of hydrocarbon emissions that contribute to the formation of smog, would be virtually eliminated.

APPLICATION TECHNIQUES

The efficiency with which a paint film protects the substrate depends on the means by which it was applied and the type of coating. The trend emerging within the industry is not only to develop improved coatings but also to develop new techniques of application aimed at improving film durability.

Probably more paint has been applied by spraying techniques than by any other method. Dipping is also a fast technique for applying coatings in a continuous operation, but it is a difficult process because of the many variables that must be closely controlled, such as viscosity and rate of withdrawal of the object being coated. Flow coating is another means of applying paint that is extensively used in industrial applications for automobile frames, appliance primers, etc. Flow coating is actually a modified form of dipping except that it has the advantage of not requiring a dip tank. Instead, the paint is sprayed on the object as it moves on a conveyor line. The problems associated with both dipping and spraying techniques include maintaining uniformity of the coatings and controlling the emission of dangerous organic solvents during application and drying of the finish. Improvements in these techniques, such as electrostatic and airless spraying, are being explored by industrial painting operations.

Other new techniques for applying and curing paints are being sought by the industry. Application of paint by electrodeposition and coil coating, and electron-beam curing of these coatings are discussed below, for they represent new technology for applying and curing modern-day coatings.

Electrodeposition

The electrodeposition process of applying paint is not a new development. A patent was obtained by Grosse and Blackwell in the 1930's for the anodic deposition of an oleoresinous coating on cans.[7] It was not until the 1950's, however, that the Ford Motor Company began to experiment with the electrodeposition of organic materials.[8,9] The main reason for the lapse of time was the slow development of suitable water-soluble paints. The Ford

Motor Company began commercial operation of electrodeposition for automobile primers in 1963. Presently the Ford Motor Company and General Motors Corporation each have seven electrodeposition systems for priming auto parts and bodies.[3]

The electrocoating process consists of three complex reactions: electrolysis, electrophoresis, and electroosmosis, all of which are believed to occur simultaneously.[7,10,11] Basically, electrodeposition is analogous to electroplating except that the metallic object to be coated is the anode and deposited organic film acts as an insulator instead of a conductor. Because the coatings act as an electrical barrier, only a single coating 1.0 to 1.5 mils thick can be applied by this technique.[3] The coating builds up uniformly over the entire metallic surface and therefore presents a definite advantage over dipping and spraying techniques. Furthermore, electrocoating uses paint solids very efficiently, so that it requires 30 percent less paint than the dipping process.[10]

At present, PPG Industries and Glidden formulate about three-fourths of the paint used in electrocoating. A wide range of polymers, including maleinized drying oils, alkyds, epoxy esters, acrylate copolymers, and phenolic modified oils, have been formulated into water systems for electrodeposition.[3] The electrodeposition process presently consumes about $10 million of the industrial paint market. About $3.5 to 4 million of paint is currently utilized in automotive electrocoating tanks.[3] This technique has not grown as rapidly as expected, primarily for two reasons:

1. Although the process lends itself to assembly-line production and operating costs are low, installation costs are high. Electrodeposition equipment for priming automobile bodies can cost between $1.5 to 2.0 million.[3]

2. The industry needs coatings of greater durability or paints formulated with conductive solids (pigments, etc.) to allow electrocoating topcoats over the primer surface. Until such paints are available, a large number of industrial painters will not consider installing electrodeposition equipment.[10,11]

Air pollution legislation regulating hydrocarbon emissions from industrial processes could very well provide the major impetus for the electrodeposition process in the 1970's, so that this technique of applying paint could mushroom, becoming a primary coating process for automobiles, appliances, electrical equipment, toys, and metal furniture. Many in the paint industry believe that within the next 10 years electrocoating could constitute 50 percent of all industrial painting.[9]

Coil Coating

Two trends that are emerging within the paint industry are exemplified in the coil-coating industry: (1) more coatings are being applied in the factory than in the field, at the expense of trade-sale paints, mainly because labor for on-site painting costs more; and (2) thinner and more uniform coatings (1 mil) are being applied on a variety of substrates (wood and metal).[12]

Coil coating, which began with the coating of venetian blind stock, is an efficient process for continually coating and curing long, flat strips of metal. At present there are about 45 large-scale coating lines utilizing industrial finishes worth about $60 to 80 million.[3] Shipments of coil-coated steel and aluminum from 1962 through the first half of 1969 are shown in Table 7.[13] Coil coating of aluminum and steel enjoyed a growth rate in the 1960's of about 13 percent per year. In 1968, more than a million tons of steel and about half a million tons of aluminum were coil-coated. Coil-coated steel and aluminum used in building and construction accounted for about half of the annual volume sales. The mobile-home industry also uses precoated aluminum for exterior siding.[14]

Table 7. SHIPMENT OF COIL-COATED STEEL AND ALUMINUM[13]

	Shipments, 10^6 lb							
	1962	1963	1964	1965	1966	1967	1968	1969[a]
Aluminum	343	404	463	553	593	727	1,006	520
Steel	585	870	1,135	1,565	1,650	1,721	2,995	1,105
Total	928	1,274	1,598	2,118	2,243	2,448	3,001	1,625

[a] First half of year.

Table 8 lists the durability and costs of some of the organic resins[15] that are used to coat steel and aluminum in the coil-coating process. Coatings such as siliconized alkyd have shown an exterior durability greater than that of conventional coatings such as the alkyd amines. A new class of polymers called fluorocarbons should find increasing use in the coil-coating industry. These coatings have resisted degradation in accelerated weathering tests.[3] Extensive field testing on industrial structures and office buildings in varying environments is currently being conducted. The use of fluorocarbons in the coil-coating industry should open new markets in commercial construction; such coatings will probably carry 15- to 20-year guarantees.[3,16,17] Because such coatings are organic-solvent-based rather than water-based, the process of coil coating is plagued with the emission of hydrocarbons. Emissions could possibly be controlled through recycling of organic solvents.

Coil coating is probably the cheapest method for applying paint to metallic substrates. Operating costs are low and little paint is wasted. As with most new techniques, there is a high initial installation cost: a high-speed line can amount to $1 million. Currently, line speeds are rated at 200 to 300 feet per minute; however, new installations are capable of 600 feet per minute. Normally such high speeds cannot be maintained, primarily because the curing cycle requires certain minimal periods. This technical problem could very well be solved by electron-beam curing of the applied coating.[3]

Table 8. COIL-COATED ORGANIC RESINS[15]

Resins	Exterior durability		Unit finishing cost range, ¢/ft²-mil
	Pigment formula	Clear film formula	
Amine alkyd	Good	Fair	⩽0.65
Polyester	Good	Fair	0.65
Vinyl-alkyd	Fair	Fair	0.65
Solution-vinyl	Good	Poor	0.85 to 1.10
Organosol	Excellent	Poor	⩽0.65
Plastisol	Excellent	Poor	⩽0.65
Straight epoxy	Poor	Poor	1.10 to 2.50
Epoxy-ester	Poor	Poor	0.85 to 1.10
Thermoset acrylic	Good	Good	0.65
Silicone alkyd	Excellent	Good	1.10 to 2.50
Polyvinyl fluoride	Excellent	Excellent	⩾2.50
Polyvinylidene fluoride	Excellent	Excellent	⩾2.50
Polyvinyl fluoride laminate	Excellent	Fair	⩾2.50
Polyvinyl chloride laminate	Excellent	Poor	⩾2.50

Electron-Beam Curing

Since the first application of paint on a production-line basis, the coating industry has been in search of a rapid cure technique. Electron-beam curing (electrocure), which is not fully commercialized, presents an entirely new concept of curing to the paint industry. This new technique uses an electron beam to induce polymerization of specially formulated paint on a flat surface. Curing by this process reportedly results in a firmer bond between the film and substrate.[3]

Electrocuring has two advantages over conventional curing techniques: (1) it cures 200 times faster than conventional methods; and (2) it cures at room temperature. Electrocuring is best used on flat stock and is limited to curing films 10 to 15 mils thick. Present paint formulations are not readily adapted to electrocuring. Resins, however, such as polyester, acrylic, urethanes, and epoxies, have been formulated with radiation-sensitive monomers or pre-polymers. The electron beam induces the formation of free radicals that react chemically with the resin to form a continuous film. These monomers or

pre-polymers can be looked upon as crosslinking reagents or curing agents. Since virtually all of the coating is converted to protective film, hydrocarbon emissions are not a problem as they are with coil coating and other bake-on processes.[3,17]

The future of electron-beam curing certainly looks bright for application in the coil-coating industry. Coil-coating line speeds could be doubled. To develop the technique, Ford Motor Company is presently operating a pilot-scale coil-coating unit in which conventional heat ovens have been replaced with electrocuring units.[3] Although there are technical problems associated with the process and the equipment is expensive, electrocuring will probably find its place in the paint industry within the 1970's.

DETERIORATION OF EXTERIOR PAINT FILMS

The paint industry is concerned about the overall performance of their paints, whether they be applied by factory techniques or by the hands of weekend painters. Consequently, the vast majority of finishes have undergone extensive testing by the industry before they are commercialized. In the following section, the testing of paint films by manufacturers is reviewed and the damaging effects of gaseous pollutants and particulate matter on exterior paints are discussed.

INDUSTRIAL TESTING

The test most widely used throughout the paint industry is the outdoor exposure test, or "fence test." Such paint-test sites can be found across the country; Florida, with its hot and humid climatic conditions, appears to be a popular area for outdoor paint-test sites. Companies such as Sherwin-Williams, DuPont, and Desoto conduct fence testing across the United States, but small paint manufacturers must rely upon published information or must conduct only limited fence tests in their own market areas.

During the actual outdoor exposure of paint films, the coating is observed periodically and the extent of degradation is recorded. Paint films have been observed to deteriorate or fail in a variety of ways, some of the more common of which are presented in Table 9. One of these types, chalking of the exterior coating, can be controlled by proper paint formulation that involves selected pigments and binders; it may even be regarded as an added asset of the paint film in some situations, because controlling the rate of chalking can produce a self-cleaning effect in the exterior paint.

Paint producers have also established a variety of tests for determining the extent of paint failure. In general, these tests involve visual observations and measurements of adhesion and stress, weight loss, gloss, reflectance, and color change of the film. Standard tests have been developed for determining the durability or weatherability of exterior paint. The American Society for Testing and Materials (ASTM) has established several such tests for evaluating paint films; some of these are listed in Table 10.

The main concern of the paint industry is the development of exterior paints that provide films of better durability under weathering. While conducting exterior exposure studies for determining the weatherability

Preceding page blank

Table 9. TYPES OF FILM FAILURE[18]

Failure	Description
Chalking	Formation of powdery layer on surface of coating that is being eroded away
Cracking or checking	Shrinkage of the coating resulting in film rupture
Alligatoring	Film rupture resulting from the application of a brittle film over a more flexible coating
Peeling	Poor adhesion of the coat to substrate
Color fading	Reaction of binder or pigment in presence of sunlight or environment
Blistering	Projections or pimples on film that result from trapping of solvent or moisture between substrate and film
Rusting	Oxidation of metallic substrates such as iron or steel when film permits moisture or chemicals to attack substrates

potential of paints, the industry studies indirectly the effects of air pollutants on exterior films; however, there has been little attempt to identify these pollutant effects. At present the paint industry is more concerned over the hydrocarbons emitted during the application of paints rather than over air pollution damage to paint films.

With the earlier paints that were formulated with oleoresinous and alkyd resins, film durability could be determined in a relatively short period of time by outdoor exposure. As formulations and new techniques have improved, paint films of greater durability have emerged on the market. Table 11 gives the expected durability of various resins used as paint binders.

The biggest drawback to the outdoor testing of organic resin topcoats with 5-, 10-, or 20-year durability is obviously the time required to obtain valid information. It seems impractical to conduct outdoor exposure studies of paints with expected durability shown in Table 11.[17] In order, then, to obtain information on the durability of such finishes, paint formulators are now using accelerated-exposure chambers. These chambers, however, are not likely to replace outdoor testing, which is regarded by the industry as the most reliable method for testing actual film durability. This report will not review the number of commercially available exposure chambers that are equipped with

Table 10. ASTM TEST METHODS FOR EXTERIOR PAINT FILMS[19]

Test no.	Test
D 659-44	Evaluating degree of resistance to chalking of exterior paints
D 660-44	Evaluating degree of resistance to checking of exterior paints
D 661-44	Evaluating degree of resistance to cracking of exterior paints
D 662-44	Evaluating degree of resistance to erosion of exterior paints
D 772-47	Evaluating degree of resistance to flaking (scaling) of exterior paints
D 1641-59	Measuring exterior durability of varnishes
D 1543-63	Measuring color change of white architectural enamels
D 1654-61	Evaluating painted or coated specimens subjected to corrosive environments
D 2197-68	Measuring adhesion of organic coatings
D 2370-68	Measuring elongation and tensile strength of free films of paint, varnish, lacquer, and related products with tensile testing apparatus.

different light sources and temperature-humidity programs; a literature survey covering the period 1956 to 1966, "Accelerated Testing of Finishes for Resistance to Weathering,"[20] showed, however, that the durability potential of paint is best predicted by the use of weatherometers equipped with sunshine-arc or xenon light sources in addition to light-dark and hot-cold-wet cycles. The survey showed also that measurements of elongation and tensile strength of the paint film are useful in predicting overall durability. Another excellent review, conducted by Battelle Memorial Institute, covered the period 1921 to 1967.[21]

The paint industry is seeking accelerated test methods that predict long-term weatherability or durability of paints. It must be pointed out that "weatherability" in reference to chamber testing does not include resistance to air pollutants as it does in reference to actual outdoor exposure tests. Although

Table 11. DURABILITY OF ORGANIC RESIN TOPCOATS [17]

Topcoat	Durability, years[a]
Oleoresinous	1 to 2
Nitrocellulose lacquer	2
Alkyd, amine	5
Acrylic lacquer	8
Acrylic enamel	8
Vinyl solution	8
Epoxy ester	2 to 5
Urethanes	2 to 10
Silicone alkyds	8 to 15
Acrylics	8 to 15
Fluoropolymers	20

[a] Years of satisfactory exposure expected for quality white formulation.

pollutant chambers are commercially available, most accelerated tests of coatings include only humidity, light, and dew cycles.

For the past several years, the National Paint, Varnish, and Lacquer Association has supported a research project at the Illinois Institute of Technology Research Institute to develop a laboratory test to be used in predicting the performance of exterior paints.[22] In this project a series of oil, alkyd, and emulsion paint films with tensile loads of 4 to 7 pounds per square inch were exposed to approximately 6 percent ozone at 35° and 60°C. The deformation of the films was recorded during periods of 165 and 505 hours of exposure to ozone. In general, for the 505-hour exposure, paints rated as first-quality have less reduction in creep compliance than second-quality paints. Figures 1 and 2 show the creep behavior of two emulsion paints: EM 7338, first-quality, and EM 7339, second-quality. There is a definite reduction in creep compliance as a result of exposure to ozone with both emulsion paints; however, the extent of reduction is about three times greater for the second-quality emulsion paint. The film of the first-quality paint was reported to retain its flexibility better than that of the second-quality emulsion.

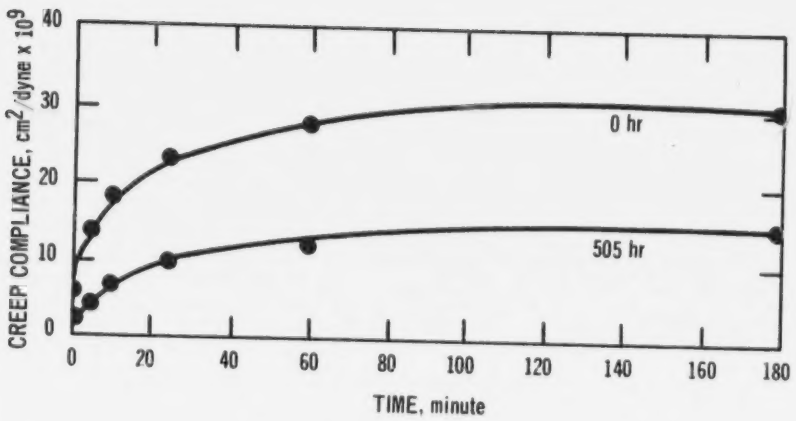


Figure 1. Initial creep of EM 7339 second-quality emulsion-base paint, before and after 505-hr exposure to ozone at 60 °C.

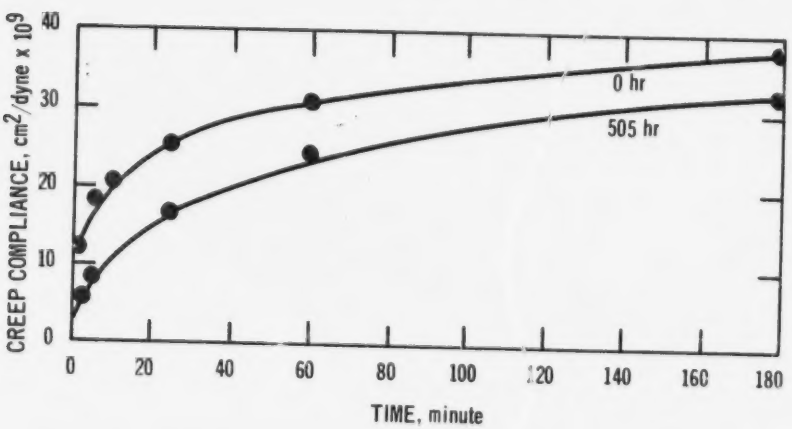


Figure 2. Initial creep of EM 7338 first-quality emulsion-base paint, before and after 505-hr exposure to ozone at 60 °C.

Ozone was used in this investigation because it is known to degrade materials by bond scission and crosslinking of the basic polymeric structure.[23] High concentrations of ozone were selected to accelerate weathering of paint films. Although films were observed to degrade in this environment, the extent that this method will be used by the industry is questionable. Such high concentrations of ozone pose a toxicity hazard in the laboratory. Also, laboratory test methods in which only one parameter is accelerated–for example, concentrations of ozone–are not likely to provide results that correlate with actual outdoor conditions.

Although the IIT Research Institute study was not initiated to investigate the effects of air pollutants on paint film, it can be looked upon as an initial investigation of the effects of a common pollutant on paints. This work should be expanded to include other gaseous pollutants, but at ambient concentrations.

EFFECTS OF AIR POLLUTION ON EXTERIOR PAINT

It has been estimated that over 213 million tons[24] of air pollutants are released annually over the United States. Primary gaseous pollutants produced

Table 12. MEAN CONCENTRATIONS OF THREE ATMOSPHERIC POLLUTANTS, 1967
($\mu g/m^3$)[a]

City	SO_2	NO_2	Total oxidants[b]
Chicago	327.5	90.0	56.8
Cincinnati	c	52.6	60.8
Denver	13.1	69.6	c
Philadelphia	256.8	80.8	51.0
St. Louis	76.0	45.1	68.6
Washington	c	80.8	49.0

[a] Data furnished by EPA Air Surveillance Network and reported in ppm.[25]
Conversion at 25°C, 760 mm Hg:
SO_2 : 1 ppm = 2620 $\mu g/m^3$
NO_2 : 1 ppm = 1880 $\mu g/m^3$
O_3 : 1 ppm = 1960 $\mu g/m^3$
[b] Total oxidants computed as ozone.
[c] Average not calculated; insufficient data.

by man are, by greatest weight, carbon monoxide, sulfur oxides, hydrocarbons, and nitrogen oxides. Primary gaseous pollutants amount to 185 million tons while the remaining 28 million tons consist of particulate matter. Not represented in these figures are localized pollutants such as hydrogen sulfide and fluoride, as well as secondary gaseous pollutants. Total amounts of the secondary pollutants that are produced from photochemical reactions of primary pollutants have been determined in certain localities but not on a national scale.

To obtain concentration data for atmospheric pollutants, the Environmental Protection Agency has placed continuous air monitoring stations in several cities of the United States. The mean concentrations of sulfur dioxide, nitrogen dioxide, and total oxidants in six cities during 1967 are shown in Table 12. The concentrations of these gaseous pollutants were found to vary considerably in each city. Of the six cities, Chicago was found to have the highest mean concentration of sulfur dioxide and nitrogen dioxide, whereas St. Louis had the highest concentration of total oxidants.

Midwest Research Institute[26] recently conducted a program in which the potential economic losses attributable to air pollution of 53 consumer materials were ranked using a systems analysis approach. The 53 consumer materials have been grouped by category in Table 13 to show the relative orders of pollution damage and available information.

The potential economic loss for damaged paints was found to hold second rank. Equally important is the fact that little has been published on the effects of air pollutants on paint finishes. The 1-year study uncovered only 11 useful references. From the Midwest Research Institute Report it is apparent that of

Table 13. RELATIVE POLLUTION DAMAGE TO MATERIALS AND AVAILABILITY OF USEFUL INFORMATION

Materials	Dollar loss, % of total	References with useful information, % of total
Metals	39	54
Paints	32	7
Textiles	9	14
Elastomers	5	12
Plastics	3	9
All others	12	4

the primary pollutants sulfur dioxide and particulate matter play an important role in the chemical deterioration of modern-day exterior paints.

Sulfur Dioxide

Sulfur dioxide has been shown to affect certain paint films at ambient concentrations under laboratory conditions. Holbrow[27] found that the drying time of linseed, tung, and bodied dehydrated castor oil paint films increased by 50 to 100 percent at exposures of 1 to 2 ppm SO_2. The touch and hard-dry times of alkyd and oleoresinous paints with titanium dioxide pigments were also reported to increase substantially. The time of exposure of the wet films to SO_2 was not reported. Analysis of the dried films indicated, however, that SO_2 had chemically reacted with the drying oils, thereby altering the oxidation-polymerization process. Based on these results, it was concluded that concentrations of SO_2 encountered in fogs near industrial sites can increase the drying and hardening times of certain kinds of paint systems.

There have been no studies reported on the effects of SO_2 on the drying of latex paints, which have captured more than 50 percent of the present exterior household market. At the time of application, SO_2 may interfere with the evaporation of water and, therefore, the coalescence of the polymeric-pigment particles. The equilibrium reaction of SO_2 and water in the coating forms sulfurous acid, which could cause instability of the protective colloid around latex particles and, consequently, flocculation. Poor film formation would be possible with resulting loss of aesthetic as well as protective value.

Holbrow also studied the effects of sulfur dioxide on the gloss of alkyd paint film.[27] In these experiments, paint films were allowed to dry, were refrigerated, and then were desiccated for 15 minutes in an atmosphere containing 1.2 percent sulfur dioxide. The paint films with condensed moisture were finally placed in an accelerated-weathering chamber. Except for a pentaerythritol alkyd paint, the gloss decreased significantly after 1 day in the accelerated-weathering chamber. Without the accelerated weathering, the actions of sulfur dioxide and moisture on the paint films produced only a slight reduction in gloss. Microscopic examination of the accelerated-weathered paint film revealed very fine wrinkles in the film. Holbrow concluded that the sulfur dioxide had sensitized the film, which permitted water to be absorbed during the weathering cycle. Control samples of paint, exposed for 1 hour of accelerated weathering, showed no loss of gloss.

Sulfur dioxide has been reported to undergo oxidative reactions in the atmosphere to produce acidic aerosols.[28] Both photochemical and catalytic reactions for this oxidization process have been investigated in many laboratories. Although the exact mechanism is unknown, sulfuric acid has been identified as a major product.[29] The deposition of sulfuric acid on painted surfaces may promote film deterioration through the reaction of the acid with various paint ingredients such as pigments, fungicides, and the binder itself. Furthermore, this acidic mist may also affect adhesion of the film on metallic substrates by promoting the occurrence of electrochemical reactions at the coating-substrate interface. As a result of these reactions, the service life of exterior paint films could be drastically reduced from that of the manufacturer's warranty.

Holbrow did not attempt to correlate moisture and pollutant concentration, or to obtain dose-response data. Although he used high levels of sulfur dioxide, his experimentation does indicate that condensation and moisture evaporation are critical in concentrating the pollutant on the surface of exterior paint films; under these conditions, deterioration of the film occurs.

Hydrogen Sulfide

Any industrial operation that liberates sulfur dioxide can be a potential source of hydrogen sulfide. Paper mills, petroleum refineries, and coke-processing plants are likely sources of hydrogen sulfide. It can also be emitted from sewage treatment plants by bacterial action on sulfur-containing organic compounds.

Hydrogen sulfide is one pollutant that attacks in-service exterior house paints. In one episode, which occurred in the southern portion of the San Francisco Bay area, house-paint darkening ranged from various tones of battleship gray to jet black. The discoloration occurred about doors and windows, under eaves, or in locations that tended to remain moist. The most severe episodes of discoloration occurred during the winter months, even though the maximum 2-hour average air concentration of hydrogen sulfide was twice as high in the summer months.[30] This phenomenon tends to confirm the finding that the presence of high humidity increases damage potentials.

The actual atmospheric concentration or threshold level of hydrogen sulfide at which paint begins to darken is unknown. From the Bay Area episodes, it appears that moisture on the paint films promotes the darkening effect. Sensenbaugh and Hemeon[31] and Sullivan[32] report that under proper conditions paint darkening is possible when the hydrogen sulfide concentration is such that the gas can be smelled, which is approximately 70 to 140 $\mu g/m^3$.

In the laboratory studies conducted by G. B. Ward,[33] it was concluded that no blackening of the films by H_2S occurred unless the paint films were actually wet with water. Unfortunately, no studies from which dose-response data can be obtained were conducted on the effects of relative humidity and H_2S interactions on paint film. The problem was traced to the formation of dark-colored metal sulfide by the chemical reaction of H_2S on lead additives and on organometallic driers and preservatives. A likely chemical reaction is:

$$H_2S + R\text{-}\overset{\overset{O}{\|}}{C}\text{-}OX \xrightarrow{H_2O} R\text{-}\overset{\overset{O}{\|}}{C}\text{-}OH + XS$$

Hydrogen sulfide + Drier → Acid + Dark salt

Mildewing of exterior paints has at times been confused with hydrogen sulfide discoloration. Mildew has been reported as the darkening of paints caused by a fungus, *Pullularia pullulans*.[30] However, many different fungi, such as *Aspergillus niger* and some species of *Alternaria* and *Cladosporium*, have been identified on coatings.[34] The fungi subsist on the organic matter or resin in the paint film. Chemical tests and microscopic techniques have been developed to differentiate between fungus attack and hydrogen sulfide discoloration of paint.

Paint producers have formulated "fume-resistant paints" that contain ingredients not susceptible to hydrogen sulfide attack. Such paints were formulated by trial-and-error procedures involving laboratory and outdoor exposures. Most exterior latex paints sold today are fume-resistant.

Particulate Matter

Exterior paints are soiled by liquids and solid particles that are composed of soot, tarry acids, and other matter of varying chemical nature. Such an agglomeration, called particulate matter, affects exterior paints in two ways: (1) loss of aesthetic attractiveness, and (2) chemical degradation of the film.

In an investigation reported by Meller, fences were erected in various areas of the United States.[35] Paint applied to the clean fences became blackened at most test sites by soot deposits. Examination of the coatings revealed that tarry matter was imbedded in the film. Efforts to remove the soiling or to clean the fence destroyed the paint film. Furthermore, Meller reported that the life of two coats of paint used to cover the soiled paint was shortened, and appearance of the paint was marred.

There have been numerous incidences of automotive paint damage near industrial process plants. Airborne iron particles from a metal-grinding operation were reported to cause pitting and staining of automobile paints.[36] Another incident was reported in which automobile paints were severely damaged by alkali mortar dust from demolition of brick buildings.[37] The vehicles in both situations had to be repainted.

Holbrow had conducted accelerated weathering tests in which various paints were exposed to particulate matter.[27] Severe staining of the paint films was observed with 0.5 ppm copper and with 0.1 ppm of iron salts. Holbrow concluded from his investigation that such staining could occur during natural exposure inasmuch as these two metals have been identified in airborne particulate matter found in United States cities and in rainwater.[38]

Data from case histories and experimental investigations show that particulate matter has been found imbedded in the paint film. Imbedded particulates can: (1) damage the coating in a physical sense; (2) provide nucleation sites at which other pollutants can concentrate; or (3) undergo chemical reactions with the paint film. In essence, particulate matter serves to

promote the chemical deterioration of exterior paints. Because there are many materials classified as particulate matter, their respective mechanisms of attack on paints are too numerous to be discussed in this report.

A program was conducted by Michelson and Tourin to investigate the frequency of house repainting as a function of suspended particulate concentrations.[39] Questionnaires were sent to residents of three suburbs of Washington, D. C. (Suitland, Rockville, and Fairfax), and two cities (Steubenville and Uniontown) in the Upper Ohio Valley. The intervals for exterior repainting as a function of particulate concentrations for the five U. S. cities, as compiled from the questionnaires, are shown in Table 14. The maintenance intervals of the years between repaintings were observed to decrease as the particulate concentrations increased. Repainting in Steubenville, which has particulate concentrations as high as 235 $\mu g/m^3$, was observed to occur about every year. In Fairfax, with a concentration of 60 $\mu g/m^3$, repainting occurred every 4 years.

Table 14. INTERVAL FOR EXTERIOR REPAINTING AS A FUNCTION OF PARTICULATE CONCENTRATION IN FIVE U. S. CITIES[39]

	Steubenville	Uniontown	Suitland	Rockville	Fairfax
Particulate concentration, $\mu g/m^3$	235	115	85	75	60
Maintenance interval, yr	0.88	1.89	2.93	3.62	3.90
Maintenance frequency, number per yr[a]	1.14	0.53	0.34	0.28	0.26

[a] Reciprocal of maintenance interval in years.

The maintenance frequency (reciprocal of maintenance interval), as a function of local particulate concentration, increases as particulate concentration increases (Figure 3). The results of the Michelson investigation are highly suggestive that a significant relationship exists between the frequency of repainting and the particulate concentration. However, extrapolation of the line indicates that repainting does not occur at zero particulate concentration and that repainting frequency continues to increase as particulate concentration increases. A leveling out of the line to form a plateau at the low and high extremes of particulate concentration should be expected. It would appear that additional maintenance data is needed, particularly for cities with mean annual particulate concentrations greater than 150 $\mu g/m^3$, before attempting to establish a more realistic relationship.

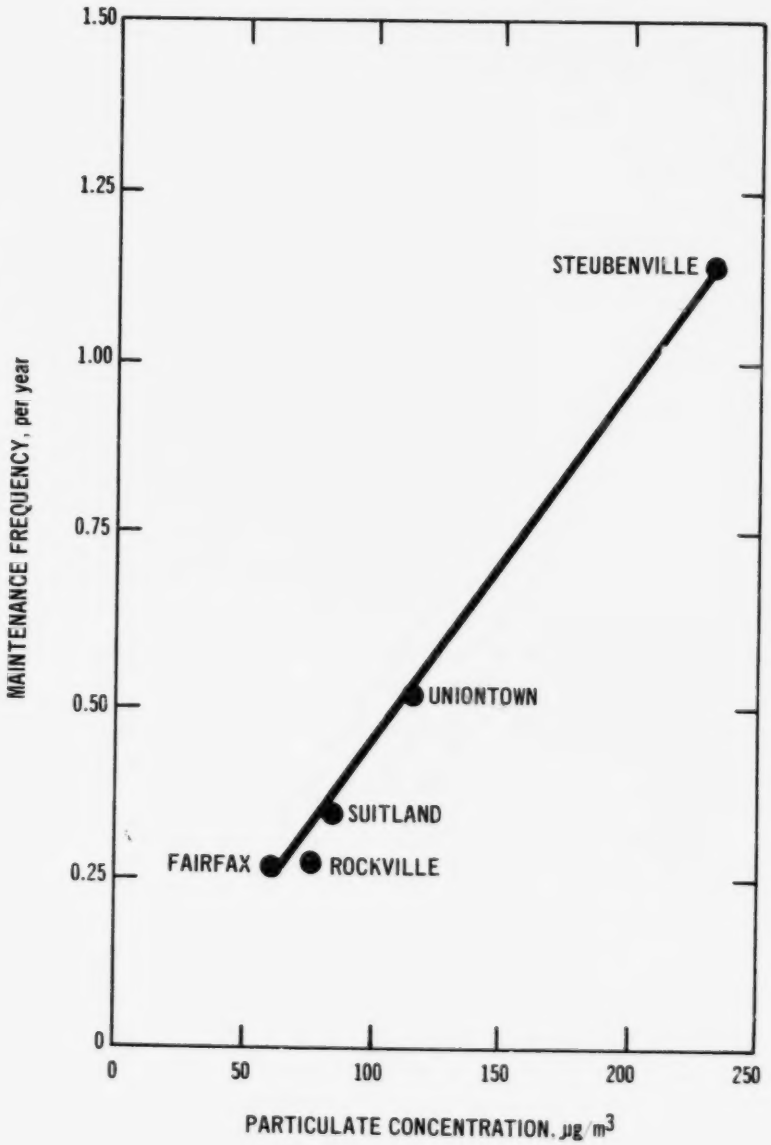


Figure 3. Relationship of maintenance frequency for exterior repainting to particulate concentration.

A study[40] to determine maintenance frequencies at various particulate concentration zones in the Philadelphia area was recently completed. In this study, problems encountered in Michelson's study were minimized, such as different characteristics in population, climate, and type of industry. The range of mean annual particulate concentrations for the Philadelphia area was 46 to 151 $\mu g/m^3$. Maintenance frequencies for exterior wall painting were obtained as shown in Table 15. This study, conducted by Booz, Allen, and Hamilton, inc., for EPA, also delineated socioeconomic factors by pollution zone. The fact that the percentage of households with incomes of less than $6000 increased with pollution level is a factor that tends to counteract the effect of suspended particulates on paint life and that partially explains why no statistically significant difference in painting frequency as a function of particulate level was detected.

Table 15. MEAN ANNUAL FREQUENCY FOR EXTERIOR WALL PAINTING IN PHILADELPHIA AREA AS A FUNCTION OF PARTICULATE CONCENTRATION

Particulate concentration ranges, $\mu g/m^3$	Exterior wall painting	
	Mean annual frequency	Standard error of mean
< 75	0.28	0.016
75 to 100	0.35	0.053
100 to 125	0.35	0.041
> 125	0.29	0.055

ECONOMIC ASSESSMENT OF AIR POLLUTION DAMAGE TO EXTERIOR PAINTS

Previous estimates of the cost of pollutant damage to exterior paints have been undertaken primarily by local government officials. Damage estimates have been basically derived from questionnaires and interviews with the general public and with paint contractors and other members of the paint industry. Although such estimates are helpful, they seldom present effects data and therefore represent an incomplete picture of the deterioration of paints caused by air pollution.

An attempt was made to establish a reasonable estimate of the potential annual consumer cost for repainting that can be attributed to pollutant damage in urban areas. This estimate, which should rank market classes by damage cost, will be used to select classes of paints for laboratory and field exposure studies. The cost estimates of the pollutant damage to exterior paints in urban areas are summarized in Table 16. The dollar volume of the 1968 paint market for four classes of exterior paints was used.

The total value at the manufacturer's level for the four paint classes was $775 million. This figure represents the dollar volume of paints used as exterior finishes and amounts to more than 50 percent of the entire 1968 paint market volume at the manufacturer's cost level. Since coil coating and maintenance finishes are not entirely exposed to the environment, the dollar volume as shown in the table is for exterior exposures only. Factory-applied automotive paints, such as the thermoplastic-thermosetting acrylic paints, are reported to protect the vehicle for the expected body life. Repainting would probably be done to repair accidental damage or to increase resale value. Therefore, only automotive refinishing paints are considered in this estimate. Of the four classes, household paints that include water- and oil-base paints comprised the largest dollar volume.

There has been no attempt to factor-out from the $775 million (column 2) the dollar volume of exterior paints that are applied to new surfaces. This damage estimate is based solely on the dollar volume of four classes of exterior paints. Whether these paints were applied to new construction or used to repaint existing structures was not considered.

In columns 3, 4, and 5 of Table 16, the expected service life and maintenance frequencies are determined for rural and urban areas for each of

Preceding page blank

Table 16. ECONOMIC ASSESSMENT OF AIR POLLUTION DETERIORATION OF EXTERIOR PAINTS

Column 1	Column 2	Column 3	Column 4	Column 5	Column 6	Column 7	Column 8	Column 9	Column 10	Column 11	Column 12
Exterior paint class	Value at manufacturer's level, $ million	Area	Expected service life, yr	Maintenance frequency, number per yr[a]	Estimated distribution of paint, % population	Paint consumed in urban areas, %	Value of paint exposed in urban areas, $ million	Service life, % loss	Loss at manufacturer's level, $ million	Labor factor	Loss at consumer's level, $ million
Coil coating	40	Rural	20	0.05	30	77	31	25	8	2	16
		Urban	15	0.07	70						
Automotive refinishing	150	Rural	5	0.20	30	74	111	20	22	4	88
		Urban	4	0.25	70						
Maintenance	100	Rural	5	0.20	30	74	74	20	15	4	60
		Urban	4	0.25	70						
Household	485	Rural	6	0.17	40	74	359	50	180	3	540
	—	Urban	3	0.33	60		—		—		—
Total	775	...	...	...	...	...	575	...	225	...	704

[a]Maintenance frequency is the reciprocal of expected service (Column 4).

the four paint classes. The average particulate concentrations were 110 $\mu g/m^3$ in U. S. cities and 40 $\mu g/m^3$ in rural areas.[41] The Michelson linear relationship, Figure 3, predicts service life of 8 and 2 years, respectively, for paint exposed to 40 and 110 $\mu g/m^3$ of particulate matter. The Booz, Allen, and Hamilton, Inc., study shows no difference in service life for paint exposed over this concentration range, the average paint life being 3.14 years. Because neither study was complete in its analysis, paint lives are estimated at 6 and 3 years, respectively, for rural and urban areas. The expected service life and the maintenance frequencies for coil coating, automotive refinishing, and maintenance coatings for rural and urban areas are not published and can only be estimated. Actual exposure data of paints exist within the paint industry, but not in terms of rural and urban areas relative to type of pollution or pollutant concentration.

Column 7 represents the amount of paint, expressed as the percentage, for each exterior paint class that is consumed in urban areas. These percentages were derived from data on maintenance frequency and estimated distribution. An example of the calculation is shown for coil coating paints:

	Maintenance frequency		Estimated distribution	
Rural area:	0.05	x	0.30	= 0.015
Urban area:	0.07	x	0.70	= 0.049
				0.064

Percentage of paint consumed in urban area:

$$\frac{0.049}{0.064} \times 100 = 77$$

As shown in column 8, paint equivalent to a total dollar volume of $575 million is exposed in urban areas. It is apparent that the major portion of this dollar volume is related to household paints. Column 8 represents the dollar volume of each paint class used annually in urban areas.

A total dollar volume of $225 million, as shown in column 10, was determined as the damage loss at the manufacturer's cost level. This damage was obtained as the product of the value of paint exposed in urban areas (column 8) and the percentage service-life loss (column 9). The percentage service-life loss attributed to air pollution was determined as the difference in expected service life between rural and urban areas. The computation shown for coil-coating, using a service life in rural areas of 20 years and a service life in urban areas of 15 years, yields the percentage service life loss resulting from air pollution:

$$\frac{20 - 15}{20} \times 100 = 25$$

The values in column 10 represent the dollar loss at the manufacturer's level rather than the retailer's. In general, the cost of paint is about one cent per square foot, or about 5 to 10 percent of the paint cost. Thus, the cost of paint represents a small portion of the total economic loss. As a conservative estimate, the mark-up, including labor cost, is over three times the value lost at the manufacturing level.[42] Therefore, the value lost at the retail level, shown in column 12 for the four classes, amounts to $704 million. Household paints, with a value loss of $540 million, represent over 75 percent of the total. A considerable portion of exterior painting, especially household, is accomplished by weekend or non-professional painters. Since a labor factor for such painters can not be determined, this dollar loss has been determined on the basis of professional painting costs.

The magnitude of error for the dollar loss of over 0.7 billion can not be determined. Before a more reliable economic assessment can be made, dose-response and expected service-life data must be obtained. Such data can only be compiled through laboratory and field studies that correlate pollutant damage with loss of service life.

AREAS FOR FUTURE INVESTIGATION

One of the important purposes of this report is to identify areas of inadequate information. The identification of information gaps will provide a starting point for further laboratory and field investigations. In summary, information is inadequate in the following areas:

1. Little information has been compiled concerning the degradation of exterior finishes by air pollutants. Although new coatings are emerging on the market at a fast pace, the following generic types of exterior paints should be considered for laboratory and field exposure studies.

   a. Household: water-base acrylic (latex) and oil-alkyd.

   b. Coil coating: urea-alkyd-coated aluminum, steel, and galvanized steel.

   c. Maintenance coating: alkyd on primed galvanized steel.

   d. Automotive finishes: nitrocellulose/acrylic.

2. Numerous test methods currently exist for assessing the weatherability of exterior paints. A program should be initiated, however, to establish test methods that correlate detectable initial film damage with service life. Such methods would greatly facilitate the study of air pollutant effects on exterior paints.

3. Environmental studies should be conducted in the laboratory to duplicate initial paint film deterioration observed in actual outdoor exposure. These laboratory studies should include: (1) dew cycle, to allow pollutants to collect and concentrate on film; (2) ultraviolet light (<2800 Å) for promoting chemical reactions; and (3) air pollutants such as ozone, sulfur dioxide, and nitrogen oxides, and particulate matter at ambient concentrations.

4. Surveys should be made of city residents and paint contractors to obtain information about the frequency of cleaning and repainting, cost of earlier repainting (labor), etc. Such information, together with effects or dose-response data, is necessary before a more realistic economic assessment of the problem can be made.

As part of an EPA effort to overcome the lack of essential information – particularly the lack of effects and dose-response data–about air pollutant damage to exterior paints, a research program for EPA was recently initiated

with the Sherwin-Williams Company.[42] This program is designed to determine the best techniques for assessing initial degradation of exterior paints, which in turn can be correlated with actual service life. The effects of ozone and sulfur dioxide at ambient concentrations on selected exterior paints will be of primary concern. Erosion, surface roughness, gloss, and tensile measurements will be evaluated as possible methods for detecting film damage. The scanning electron microscope will provide a visual record of topographical changes. Techniques which are developed in this program will be used at EPA's laboratory facilities at Research Triangle Park, North Carolina. Both community and controlled environment studies are planned in our programs to determine the effects of SO_2, NO_2, and ozone on consumer materials.

CONCLUSIONS

In order to compete with the industrial market demand for structural materials that require little or no painting, the paint industry is beginning to develop new paint formulations. Current technology is centered around the development of synthetic resins and of new techniques for applying and curing these paints. Since hydrocarbons are emitted during the use of some of these new techniques, extant and proposed air pollution legislation that regulates hydrocarbon emissions will affect this technology. Such legislation should give impetus to the formulation and use of new water-base industrial paints or paints formulated with hydrocarbon solvents that are not photochemically reactive.

A review of literature on the chemical attack of certain air pollutants on exterior finishes reveals that dose-response data for the effects of common air pollutants on exterior paints are sadly lacking at this time. It is known, however, that ozone, sulfur dioxide, hydrogen sulfide, and particulate matter can cause damage to exterior paints. Only a few mechanisms of chemical attack of pollutants on paints have as yet been elucidated and these are noted in this report.

An economic assessment of the damage caused by air pollutants to exterior paints reveals that the cost at the consumer level of such damage is more than $0.7 billion annually. Of the four categories of exterior paints studied—household, automotive refinishing, coil coating, and maintenance—household paints sustain damage representing over 75 percent of the total annual dollar loss.

Because dose-response data on the effects of common air pollutants on exterior paints are lacking, future research should be directed toward remedying this deficiency. Field and laboratory studies of the degradation of exterior finishes by air pollutants should be conducted. In addition, test methods should be developed for correlating paint film damage with service life. Exposure-chamber studies should be conducted on the effects of common air pollutants on paints. In addition to these studies, surveys of city residents and paint contractors should be made to obtain information on maintenance repainting. Only when survey and effects or dose-response data are available can a realistic economic appraisal of the problem be made.

REFERENCES

1. Noble, P. *Marketing Guide to the Paint Industry.* Fairfield, New Jersey, Charles H. Kline & Co., Inc., 1969.

2. Martens, C. R. *Technology of Paints, Varnishes, and Lacquers.* New Jersey, Reinhold Book Corporation, 1968.

3. Kiefer, D. M. The Paint Industry. Chem. Eng. News. *47*(53):32-57, December 1969.

4. Banor, Abel. Industrial Finishes Gallonage Due to Surpass Trade Sales in the Soaring Seventies. Amer. Paint J., Convention Daily:8-9, October 27, 1970.

5. National Paint, Varnish, and Lacquer Association Circular 799, April 1970.

6. Banor, Abel. Coatings Industry Projections: Who Will Gain or Lose - Raw Material Tonnage - 1970-75. Amer. Paint J., Convention Daily:16-20, October 29, 1969.

7. Curtis, W. B. *et al.* A Survey of the Present Theory and Practice of the Application of Paint by Electrodepo: tion Process. Chem. and Ind. 1858-1862, November 7, 1967.

8. Brewer, G.E.F. *et al.* Ford Electrocoating Process. J. Paint Technol. *39*(512):551-553, September 1967.

9. Burnside, G. L. Ford Electrocoating Process: Principles of Process, Feed, and Engineering. Mater. Prot. 37-39, October 1968.

10. Kock, Erhard. Electrodeposition Organic Coatings. Mater. Des. Eng. 70-75, October 1966.

11. Hays, D. R., and C. S. White. Electrodeposition of Paint: Deposition Parameters. J. Paint Technol. *41*(535):461-471, August 1969.

12. Oliver, J. What's Ahead In Coatings for Products Finishing. Prod. Finish. 40-46 August 1968.

13. Data supplied by National Coaters Association, 1900 Arch Street, Philadelphia, Pa

Preceding page blank

14. Walker, P. Steel Comeback Predicted in Cans and Mobile Homes. Metal Work News. October 1969.

15. Mock, J. A. A Guide to Prepainted Metal Finishes. Mater. Des. Eng. 89-92, November 1969.

16. Preuss, H. Organic (Paint) Coating, Processes, and Equipment. Metal Finish. 58-70, January 1969.

17. Mock, J. A. What's New in Organic Industrial Coating. Mater. Eng. 60-63, December 1962.

18. NACE Publication 6D170. Causes and Prevention of Coatings Failure. Mater. Protection. 32-36, March 1970.

19. Test for Formulated Products. 1970 Annual Book of ASTM Standards. Part 21: Paint, Varnish, Lacquer, and Related Products.

20. Kuenstler, H. G., and E. G. Shur. Accelerated Testing of Finishes for Resistance to Weathering. J. Paint Technol. *40*(516):48A-54A, January 1968.

21. Brand, B. G. *et al.* Predicting Service Life of Organic Coating. J. Paint Technol. *40*(524):396-425, September 1968.

22. Gutfreund, K. Determination of the Susceptibility of Paint Films to Deterioration. J. Paint Technol. *38*(503):732-739, December 1966.

23. Bailey, P. S. The Reactions of Ozone with Organic Compounds. Chem. Rev. *48*:925-1010, 1958.

24. *Nationwide Inventory of Air Pollutant Emissions - 1968*, DHEW, PHS, EHS, National Air Pollution Control Administration, Raleigh, North Carolina. August 1970.

25. Air Quality Data from the National Air Surveillance Networks. Raleigh, North Carolina. 1967 Edition, November 1969.

26. Midwest Research Institute, *Systems Analysis of the Effects of Air Pollution on Materials*, Raleigh, N. C., Final Report, Contract No. CPA-22-69-113, January 1970.

27. Holbrow, G. L. Atmospheric Pollution: Its Measurement and Some Effects on Paint. J. Oil Colour Chem. Ass. *45*:701-718, October 1962.

28. Moeller, T. *Inorganic Chemistry*. New York, John Wiley and Sons, Inc., 1963, 966p.

29. Gerhard, E. R. and G. F. Johnston, Photochemical Oxidation of Sulfur Dioxide in Air. Ind. Engr. Chem., *47*:972-976, 1955.

30. Wohlers, H. C., and M. Feldstein. Hydrogen Sulfide Darkening of Exterior Paint. J Air Pollut. Control Ass. *16*(1):19-21, January 1966.

31. Sensenbaugh, J. D., and W. C. L. Hemeon. A Low Cost Sampler for Measurement of Low Concentration of Hydrogen Sulfide. J. Air Pollut. Control Ass. *4*:5-8, May 1954.

32. Sullivan, J. L. An Investigation of Air Pollution Problems in the South Auckland Area. Wellington, New Zealand, R. E. Owens, Government Printer, 1957.

33. Ward, G. B. Blackening Effect of Hydrogen Sulfide on Exterior White House Paints. Off. Dig. 1089-1100, November 1956.

34. Ross, R. T. Microbial Deterioration of Paint Films. Developments in Industrial Microbiology. *6*:149-163,1964.

35. Meller, H. B. Damage Due to Smoke. TASME FSP-50-75.

36. Fochtman, E. G., and G. Langer. Automobile Paint Damage by Airborne Iron Particles. J. Air Pollut. Control Ass. *6*(4):243-247, February 1957.

37 Yocom, J. E. The Deterioration of Materials in Polluted Atmospheres. J. Air Pollut. Control Ass. *8*(3):203-208, November 1958.

38. Tabor, E. C., and W. V. Warren. Distribution of Certain Metals in the Atmosphere of Some American Cities. Arch. Ind. Health *17*:145-151, 1958.

39. Michelson, I., and B. Tourin, *Report on Study of Validity of Extension of Economic Effects of Air Pollution Damage from Upper Ohio River Valley to Washington, D. C. Area.* Environmental Health and Safety Research Association, Contract PH 27-68-22, August 1967.

40. Booz, Allen, and Hamilton, Inc. *Study to Determine Residential Soiling Costs of Particulate Air Pollution.* Raleigh, N. C., Final Report, Contract No. CPA 22-69-103, October 1970.

41. *Air Quality Criteria for Particulate Matter,* DHEW, PHS, National Air Pollution Control Administration, Washington, D. C., Publication No. AP-49, pp. 11-15, January 1969.

42. The Sherwin-Williams Company. Study to Evaluate Techniques for Assessing Air Pollution Damage to Paint. Air Pollution Control Office, Raleigh, North Carolina. Contract No. EHSD 71-41.

BIBLIOGRAPHY

TESTING OF PAINTS

1. Berg, C. J. *et al.* Performance of Polymers in Pigment Systems. J. Paint Technol. *39*(510):436-453, July 1967.

2. Bigos, J. *et al.* Five Year Test Results: AISI Research Project on Paintability of Galvanized Steel. J. Paint Technol. *39*(508):316-327, May 1967.

3. Brown, T. C., and L. N. Whitehill. Durability of Alkyd Amino Resin Systems Based on Accelerated and Exterior Exposure Results. J. Paint Technol. *38*(501):615-619. October 1966.

4. Dunkley, F. G., and D. P. Earp. The Correlation of Service Behavior of Paint with Observed Physical Characteristics in Air Drying Paint for Structures. J. Oil and Colour Chem. *44*(10):649-688, October 1961.

5. Gackenbach, R. E. A Laboratory Paint Test Program. Corrosion *16*(1):1t-4t, January 1960.

6. Hearst, P. J. Volatile Degradation Products of Organic Coatings Irradiated in Air. J. Paint Technol. *39*(506):119-127, March 1967.

7. Newton, D. S., and J. G. Rigg. The Exterior Durability of Paints Based on Lithopane and Zinc Sulphide. J. Oil and Colour Chem. *44*(12):835-850, December 1961.

8. Nowacki, L.J. An Evaluation of Various Weatherometers of Determining the Service Life of Organic Coating. Official Digest *37A*:1371-1391, November 1965.

9. Martin, K. E., and R. Rolles. Durability of Precoated Aluminum Products. J. Paint Technol. *39*(510):418-425, July 1967.

10. Schurr, G. G. *et al.* Possibility of Predicting Exterior Durability by Stress/Strain Measurements. J. Paint Technol. *38*(501):591-599, October 1966.

AIR POLLUTANTS AND PAINTS

1. Bwidick, L. R., and J. F. Barkley. Effects of Sulfur Compounds in the Air on Various Materials. Bureau of Mines. Circular 7064. April 1939.

2. George, W. F. Air Pollution and Protective Coatings: Houston, Dallas, and Washington. J. Paint Technol. *40*(520):222-228, May 1968.

3. Graff-Baker, C. The Effect of Dust Particles on the Electrical Resistance and Anti-Corrosive Properties of Varnish and Paint Films. J. Appl. Chem. *8*: 590-598, September 1958.

4. Hayakowa, K. Air Pollution and Protective Coatings. Japan Air Cleaning Assoc. (Tokyo) *4*(4):36-38, 1966.

5. Larsen, R. I. Air Pollution Damage. Am. Paint J. *42*(5):94-109, October 14, 1957.

6. Pitts, J. W., and D. G. Moore. Apparatus for Studying the Effects of Atmospheric Pollution and Cyclic Dew Formation on the Deterioration of Materials. Materials Research and Standard *6*(7):328, 1966.

7. Schwebmann, J. C. Natural Phenomena in Air Pollution. J. Air Pollut. Control Ass. *14*(2):48-49, February 1964.

TRENDS IN THE PAINT INDUSTRY

1. Buttignol, V. J. *et al.* Protective Coatings. Ind. and Eng. Chem. *58*(8):44-56, August 1966.

2. Levinson, S. B., and S. Sprindel. Recent Developments in Architectural and Maintenance Painting. J. Paint Technol. *40*(520):25A-33A.

3. Miranda, T. J., and T. F. Huemmer. Radiation Curing of Coatings. J. Paint Technol. *41*(529):118-127, February 1969.

4. Moore, S. *et al.* Trends in the Coatings Industry II. Am. Paint J. *55*(5):54 66, August 24, 1970.

5. Patton, T. C. Review of Fundamentals and Advances in Urethane Coating Technology. J. Paint Technol. *39*(512):554-563. September 1967.

6. Schneider, H. J. Coil Coating: Continuing Challenge to the Coatings Industry. J. Paint Technol. *40*(521):48A-52A, June 1968.

☆ U. S. GOVERNMENT PRINTING OFFICE 1972—484-486/242

ENVIRONMENTAL PROTECTION AGENCY
Office of Air Programs - OTIP
Research Triangle Park, N.C. 27711

OFFICIAL BUSINESS

POSTAGE AND FEES PAID
ENVIRONMENTAL PROTECTION AGENCY

If you do not desire to continue receiving this technical report series, please CHECK HERE. ☐ , tear off this label, and return it to the above address. Your name will then be promptly removed from the appropriate mailing list.

APR 1972

OFFICE OF AIR PROGRAMS PUBLICATION No. AP–103